

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

20-4744P

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

09/623010

INTERNATIONAL APPLICATION NO.

PCT/JP99/00825

INTERNATIONAL FILING DATE

February 24, 1999

PRIORITY DATE CLAIMED

February 27, 1998

TITLE OF INVENTION

AQUEOUS FLUROELASTOMER CURABLE COMPOSITION AND COATED ARTICLE

APPLICANT(S) FOR DO/EO/US

TOMIHASHI, Nobuyuki; OGITA, Koichiro; TERASAKA, Kiyotaro; NAKATANI, Yasukazu

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
- a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
- b. ☒ has been transmitted by the International Bureau.
- c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(3)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(2)).
- a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
- b. ☐ have been transmitted by the International Bureau.
- c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
- d. ☒ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98./International Search Report with cited references
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A **FIRST** preliminary amendment.
☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information:
1.) Zero (0) sheets of Formal Drawings
2.) Amended claims under PCT Article 34

U.S. APPLICATION NO (if known, see 37 CFR 1.5) 09/623010		INTERNATIONAL APPLICATION NO PCT/JP99/00825		ATTORNEY'S DOCKET NUMBER 20-4744P	
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<p>17. <input checked="" type="checkbox"/> The following fees are submitted:</p> <p>BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5): Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO. \$970.00</p> <p>International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$840.00</p> <p>International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO. \$690.00</p> <p>International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$670.00</p> <p>International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4). \$96.00</p> <p>ENTER APPROPRIATE BASIC FEE AMOUNT =</p> <p>Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).</p> <table border="1" style="width:100%; border-collapse: collapse;"> <tr> <th style="width:20%;">CLAIMS</th> <th style="width:20%;">NUMBER FILED</th> <th style="width:20%;">NUMBER EXTRA</th> <th style="width:20%;">RATE</th> <th style="width:20%;"></th> <th style="width:20%;"></th> </tr> <tr> <td>Total Claims</td> <td>20 - 20 =</td> <td>0</td> <td>X \$18.00</td> <td>\$</td> <td>0</td> </tr> <tr> <td>Independent Claims</td> <td>1 - 3 =</td> <td>0</td> <td>X \$78.00</td> <td>\$</td> <td>0</td> </tr> </table> <p>MULTIPLE DEPENDENT CLAIM(S) (if applicable) Yes <input checked="" type="checkbox"/> + \$260.00 \$ 260.00</p> <p>TOTAL OF ABOVE CALCULATIONS = \$ 1230.00</p> <p>Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28). \$ 0</p> <p>SUBTOTAL = \$ 1230.00</p> <p>Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)). \$ 0</p> <p>TOTAL NATIONAL FEE = \$ 1230.00</p> <p>Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property + \$ 0</p> <p>TOTAL FEES ENCLOSED = \$ 1230.00</p> <table border="1" style="width:100%; border-collapse: collapse;"> <tr> <td style="width:60%;"></td> <td style="width:20%; text-align: right;">Amount to be:</td> <td style="width:20%;"></td> </tr> <tr> <td></td> <td style="text-align: right;">refunded</td> <td>\$</td> </tr> <tr> <td></td> <td style="text-align: right;">charged</td> <td>\$</td> </tr> </table>	CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE			Total Claims	20 - 20 =	0	X \$18.00	\$	0	Independent Claims	1 - 3 =	0	X \$78.00	\$	0		Amount to be:			refunded	\$		charged	\$	<p>CALCULATIONS</p> <p>PTO USE ONLY</p>
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a. ☒ A check in the amount of \$ 1230.00 to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account. No. _____ in the amount of \$ _____ to cover the above fees.
 A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any
 overpayment to Deposit Account No. 02-2448.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

Send all correspondence to:
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SIGNATURE

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#32,868 (ADM)
REGISTRATION NUMBER

/cqc August 25, 2000

09/623010

PATENT
533 Rec'd PCT/PTO 25 AUG 2000 1744P

IN THE U.S. PATENT AND TRADEMARK OFFICE

Applicant: TOMIHASHI, Nobuyuki et al.
Int'l. Appl. No.: PCT/JP99/00825
Appl. No.: New Group:
Filed: August 25, 2000 Examiner:
For: AQUEOUS FLUOROELASTOMER CURABLE
COMPOSITION AND COATED ARTICLE

PRELIMINARY AMENDMENT

BOX PATENT APPLICATION

Assistant Commissioner for Patents
Washington, DC 20231

August 25, 2000

Sir:

The following Preliminary Amendments and Remarks are respectfully submitted in connection with the above-identified application.

AMENDMENTS

IN THE SPECIFICATION:

Please amend the specification as follows:

Before line 1, insert --This application is the national phase under 35 U.S.C. § 371 of PCT International Application No. PCT/JP99/00825 which has an International filing date of February 24, 1999, which designated the United States of America.--

IN THE CLAIMS:

Please amend the claims as follows:

Claim 10: Line 4, change "any one of claims 1 to 9" to
--claim 1--

REMARKS

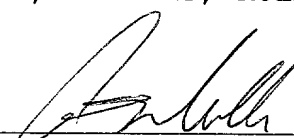
The specification has been amended to provide a cross-reference to the previously filed International Application. The claims have also been amended to delete improper multiple dependents and to place the application into better form for examination. Entry of the present amendment and favorable action on the above-identified application are respectfully requested.

If necessary, the Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 02-2448 for any additional fees required under 37 C.F.R. § 1.16 or under 37 C.F.R. § 1.17; particularly, extension of time fees.

Respectfully submitted,

BIRCH, STEWART, KOLASCH & BIRCH, LLP

By


Andrew D. Meikle, #32,868

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(Rev. 04/19/2000)

DESCRIPTION

AQUEOUS FLUOROELASTOMER CURABLE COMPOSITION AND COATED ARTICLE

FIELD OF THE INVENTION.

5 The present invention relates to an aqueous fluoroelastomer curable composition and a coated article. In particular, the present invention relates to an aqueous fluoroelastomer curable composition comprising a fluorine-containing copolymer and a specific polyol curing agent, and an article having a coating film
10 formed from such a composition.

BACKGROUND ART

 A method for curing an aqueous fluoroelastomer coating composition with a polyamine curing agent is known (see JP-B-58-53671 corresponding to US-A-4339553). In general, the coating
15 film formed by this method has good mechanical properties, but low sealing properties. In addition, when such a coating film is used to coat the surface of a roll used in office automation (OA) equipment (e.g. copying machines, printers, etc.), it has drawbacks such as insufficient elasticity, heat resistance, and
20 so on. When the polyamine curing agent is added to a fluoroelastomer curable composition, for example, a coating composition, its pot life is shortened because of gelling, etc. Accordingly, two or more fluid type compositions are practically supplied, but no practically usable one-fluid type composition
25 is available.

 When a fluoroelastomer is cured with a polyol curing agent, it is possible to prepare one-fluid type fluoroelastomer coating compositions containing organic solvents (see. PCT/JP97/02853).

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However, in these years, the VOC regulation and the like become severer and severer, and thus environment friendly aqueous coating compositions are desired. However, one-fluid type aqueous fluoroelastomer coating compositions in the form of an aqueous dispersion containing no organic solvent are not known.

DISCLOSURE OF THE INVENTION

One object of the present invention is to provide a one-fluid type aqueous fluoroelastomer curable composition, which can be formulated as an aqueous dispersion, form a cured film having high strength at a relatively low temperature in a short time, and has a long pot life.

Another object of the present invention is to provide a coated article, which does not suffer from the drawbacks of coated articles having a coating film formed from a conventional aqueous fluoroelastomer coating composition, in particular, rolls for OA equipment.

To achieve these and other objects, the present invention provides an aqueous fluoroelastomer curable composition comprising an aqueous fluoroelastomer dispersion and a basic polyol curing agent, and a coated article at least a part of the surface of which is coated with a coating layer formed from such a composition.

EMBODIMENTS TO CARRY OUT THE INVENTION

Now, the components contained in the composition of the present invention will be explained.

(A) Aqueous dispersion of fluoroelastomer

The aqueous dispersion of a fluoroelastomer is prepared by dispersing a fluorine-containing elastic copolymer in water in

a concentration of 10 to 75 wt. % in the presence of a surfactant such as polyoxyethylene, an alkyl phenyl ether, an alkylsulfonate salt, etc.

The fluorine-containing elastic copolymer is a
 5 fluorine-containing copolymer comprising repeating units represented by the formula: $-\text{CH}_2-$ in the backbone. Typical examples of such a copolymer include fluorine-containing elastic copolymers comprising vinylidene fluoride, and specific examples of such copolymers are those comprising the following repeating
 10 units in the backbone:

at least one repeating unit selected from the group consisting of $-\text{CF}_2-\text{CH}_2-$, $-\text{CH}_2-\text{CH}_2-$ and $-\text{CH}_2-\text{CH}(\text{CH}_3)-$ and

at least one repeating unit selected from the group
 15 consisting of $-\text{CF}_2-\text{CF}(\text{CF}_3)-$, $-\text{CF}_2-\text{CF}_2-$ and $-\text{CF}_2-(\text{ORf})\text{CF}-$ in which Rf is a fluoroalkyl group having 1 to 6 carbon atoms.

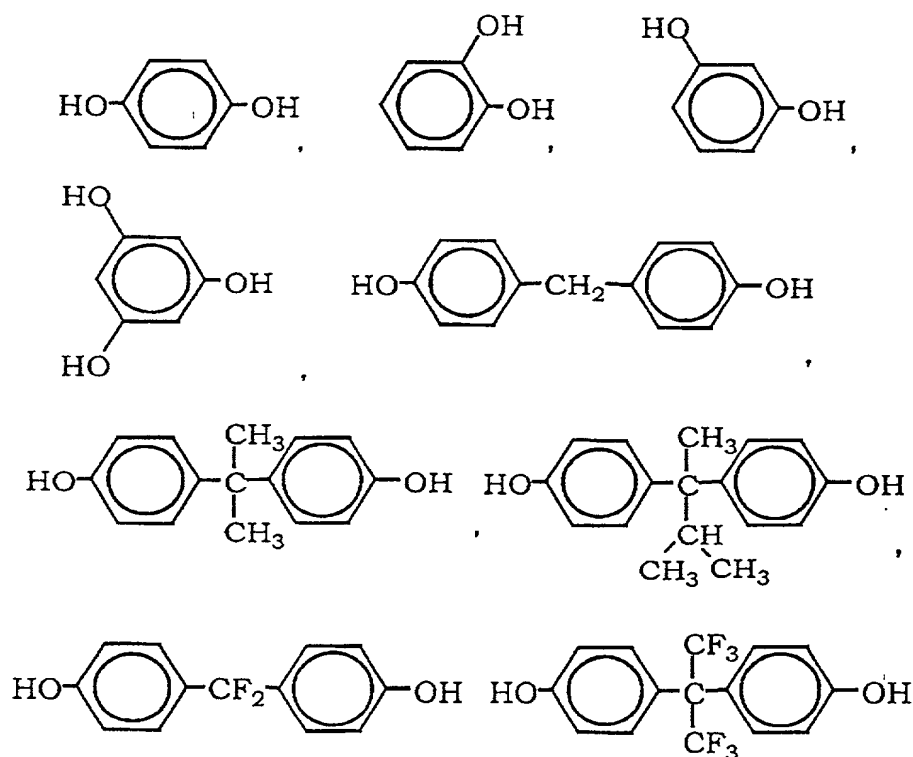
Specific examples of such copolymers include vinylidene fluoride-hexafluoropropylene copolymers, vinylidene fluoride-tetrafluoroethylene-hexafluoropropylene copolymers, ethylene-hexafluoropropylene copolymers, tetrafluoroethylene-propylene
 20 copolymers, etc. Such fluorine-containing elastic copolymers are commercially sold under the trade names of "DAIEL[®]" (by Daikin Industries Ltd.), "BAITON FLOAM[®]" (by E.I DuPont), "AFLAS[®]" (by ASAHI GLASS Co., Ltd.), and so on. Among them, the vinylidene
 25 fluoride base copolymers are preferable in view of the crosslinkability.

(B) Basic polyol curing agent

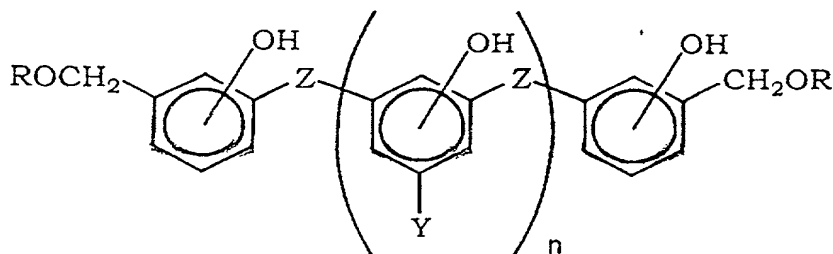
Herein, the basic polyol curing agent means a compound or

a polymeric compound having at least two hydroxyl groups, in particular, phenolic hydroxyl groups in the molecule, and the curing ability.

Examples of the polyol curing agent includes salts of a basic
5 compound with a phenol compound of the formula:



or a phenolic resin of the formula:



10 wherein Z is $-\text{CH}_2-$ or $-\text{CH}_2\text{OCH}_2-$, Y is a hydrogen atom, a halogen

atom, or a group of the formula: $-R$, $-CH_2OR$ or $-OR$ in which R is an alkyl group having 1 to 4 carbon atoms, and n is an integer of 0 to 100.

Examples of the basic compound include ammonium, 5 phosphonium, alkali metals (e.g. lithium, sodium, potassium, etc.), alkaline earth metals (e.g. beryllium, magnesium, calcium, barium, etc.), and so on.

Among them, the salts of hydroquinone, bisphenol A, bisphenol AF, resol type phenolic resins, etc. are preferable in 10 view of the properties of the coating films.

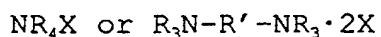
As a curing agent, a combination of the above basic polyol and other polyol may be used.

(C) Curing accelerator

The composition of the present invention may contain the 15 following compound as an optional curing accelerator:

Quaternary ammonium salt

A quaternary ammonium salt of the formula:



wherein X is an acid residue or a hydroxyl group, R groups are 20 the same or different and each an alkyl group having 1 to 20 carbon atoms, an halogenated alkyl group having 1 to 20 carbon atoms or an aryl group having 6 to 20 carbon atoms, or two or more R groups together form a carbon ring or a heterocyclic group, and R' is an alkylene group having 2 to 21 carbon atoms or a 25 phenylenedialkylene group having 8 to 12 carbon atoms.

Examples of the acid residue include halide, sulfate, sulfite, bisulfite, thiosulfate, sulfide, polysulfide, hydrogen sulfide, thiocyanate, carbonate, bicarbonate, nitrate,

carboxylate, borate, phosphate, biphosphate, phosphite,
perchlorate, bifluoride, arsenate, ferricyanide, ferrocyanide,
molybdate, selenate, selenite, uranate, tungstate, etc.

Specific examples of the quaternary ammonium salt include

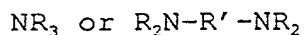
- 5 quaternary alkyl- and aralkyl-ammonium salts (e.g.
trimethylbenzylammonium chloride, triethylbenzylammonium
chloride, dimethyldecylbenzylammonium chloride,
triethylbenzylammonium chloride,
myristylbenzyltrimethylammonium chloride,
10 dodecyltrimethylammonium chloride,
dimethyltetradecylbenzylammonium chloride,
trimethyltetradecylammonium chloride, coconuttrimethylammonium
chloride, stearyltrimethylammonium chloride,
distearyldimethylammonium chloride, tetrabutylammonium
15 hydroxide, 1,4-phenylenedimethylenebis(trimethylammonium
dichloride, 1,4-phenylenedimethylenebis(triethylammonium
dichloride, ethylenebis(triethylammonium dibromide, etc.),
quaternary 1,8-diaza-bicyclo[5.4.0]-7-undecenium salts (e.g.
8-methyl-1,8-diaza-bicyclo[5.4.0]-7-undecenium chloride, 8-
20 methyl-1,8-diaza-bicyclo[5.4.0]-7-undecenium iodide, 8-
methyl-1,8-diaza-bicyclo[5.4.0]-7-undecenium hydroxide, 8-
methyl-1,8-diaza-bicyclo[5.4.0]-7-undecenium methylsulfate,
8-methyl-1,8-diaza-bicyclo[5.4.0]-7-undecenium bromide, 8-
propyl-1,8-diaza-bicyclo[5.4.0]-7-undecenium bromide, 8-
25 dodecyl-1,8-diaza-bicyclo[5.4.0]-7-undecenium chloride, 8-
dodecyl-1,8-diaza-bicyclo[5.4.0]-7-undecenium hydroxide, 8-
eicosyl-1,8-diaza-bicyclo[5.4.0]-7-undecenium chloride, 8-
tetracosyl-1,8-diaza-bicyclo[5.4.0]-7-undecenium chloride, 8-

benzyl-1,8-diaza-bicyclo[5.4.0]-7-undecenium chloride, 8-benzyl-1,8-diaza-bicyclo[5.4.0]-7-undecenium hydroxide, 8-phenethyl-1,8-diaza-bicyclo[5.4.0]-7-undecenium chloride, 8-(3-phenylpropyl)-1,8-diaza-bicyclo[5.4.0]-7-undecenium
 5 chloride, etc.), and the like.

Furthermore, as curing accelerators, quaternary salts of the following tertiary amines with inorganic or organic acids can be used:

Tertiary amine

10 A tertiary amine of the formula:



wherein R groups are the same or different and each an alkyl or alkenyl group having 1 to 20 carbon atoms, or an aryl group having 6 to 20 carbon atoms, or two or more R groups together form a carbon
 15 ring or a heterocyclic group, and R' is an alkylene group having 2 to 21 carbon atoms or a phenylenedialkylene group having 8 to 12 carbon atoms.

Specific examples of the tertiary amines include trimethylamine, triethylamine, tri-n-propylamine, tri-n-
 20 butylamine, triisobutylamine, methyldiethylamine, dimethylethylamine, dimethyl-n-propylamine, dimethyl-n-butylamine, dimethylisobutylamine, dimethylisopropylamine, dimethyl-sec.-butylamine, dimethyl-tert.-butylamine, triallylamine, diallylmethylamine, allyldimethylamine,
 25 benzyl dimethylamine, benzyl diethylamine, N-allylpiperidine, N-ethylpiperidine, N-butylpiperazine, N-methylpyrrolidine, N-cyclohexylpyrrolidine, N-n-butylpyrrolidine, N-ethylpyrrolidine, N-benzylpyrrolidine, 2,4,6-trimethylpyridine,

etc.

Examples of the inorganic or organic acids which form the quaternary salts include HCl, HBr, HF, $(C_2H_5)_3NH^+Cl^-$, $(C_2H_5)_3NH^+NO_3^-$, $2(C_2H_5)_3NH^+SO_4^{2-}$, $2(C_2H_5)_3NH^+CO_3^{2-}$, $(C_2H_5)_3NH^+RO^-$, $(C_2H_5)_3NH^+RCOO^-$,
 5 $(C_4H_9)_3NH^+Cl^-$, $(C_4H_9)_3NH^+NO_3^-$, $2(C_4H_9)_3NH^+SO_4^{2-}$, $2(C_4H_9)_3NH^+CO_3^{2-}$,
 $(C_4H_9)_3NH^+RO^-$, $(C_4H_9)_3NH^+RCOO^-$ wherein R is an alkyl or alkenyl group having 1 to 20 carbon atoms or an aryl group having 6 to 20 carbon atoms.

Primary or secondary amines are not preferable since they
 10 induce reactions other than the polyol curing. Amines having pKa of less than 8 are not preferable, since they achieve the low curing rate and thus the coating film has low strength.

In the present invention, an organic acid may be added to the composition to improve the shelf stability of the composition.
 15 As the organic acid, one having 1 to 12 carbon atoms, preferably 1 to 4 carbon atoms is used. Among them, those having 9 or more carbon atoms are less preferable since they remain in the coating film formed from the composition. Preferred organic acids are monocarboxylic acids such as formic acid, acetic acid, propionic
 20 acid, etc. and dicarboxylic acids such as oxalic acid, malonic acid, succinic acid, etc.

The organic acids suppress the polyol curing of the fluorine-containing copolymer contained in the composition during the storage. When the composition is coated, dried and baked,
 25 the organic acid is evaporated or thermally decomposed, the basic compound accelerates the curing reaction. Thus, the organic acid is categorized into "curing accelerators" in the present invention.

The compounded amounts of the components in the composition according to the present invention are as follows:

The basic polyol curing agent is used in an amount of 0.1 to 10 wt. parts, preferably 0.5 to 5 wt. parts, and the curing
5 accelerator is used in an amount of 0 to 10 wt. parts, preferably 0.01 to 5 wt. parts, all based on 100 wt. parts of the fluorine-containing copolymer.

When the amount of the curing accelerator is less than the above lower limit, it may be difficult to cure the copolymer. When
10 the amount of the curing accelerator exceeds the above upper limit, it may be difficult to control the curing.

In addition to the polyol curing agent and the curing accelerator, the composition of the present invention may contain various additives which are usually added to the fluoroelastomer
15 compositions, for example, fillers, colorants, acid-acceptors, etc.

Examples of the filler include carbon black, white carbon, calcium carbonate, barium sulfate, etc., and examples of the colorant include inorganic pigments, mixed oxide pigments, etc.

20 Examples of the acid-acceptor include magnesium oxide, lead oxide, zinc oxide, lead carbonate, zinc carbonate, and complex salts such as hydrotalcite. Acid-acceptors having a high activity such as calcium hydroxide are less preferable since the composition tends to gel. Acid-acceptors having lower pKa than
25 that of the above basic compounds are preferable. When the acid-acceptors have high pKa, the composition tends to gel. In general, the acid-acceptor is used in an amount of 1 to 40 wt. parts per 100 wt. parts of the fluorine-containing copolymer in

accordance with the activity of the acid-acceptor.

Furthermore, the composition of the present invention may contain 5 to 900 wt. parts of a fluororesin or a terminal-modified perfluoropolyether (a perfluoropolyether compound having a
5 terminal functional group reactive with the fluorine-containing copolymer such as $-NH_2$, $-CH_2OH$, etc.) per 100 wt. parts of the fluorine-containing copolymer. Thereby, the non-stick property can be imparted to the coating film formed from the composition.

Examples of the fluororesin include polyvinylidene
10 fluoride (PVdF), ethylene-tetrafluoroethylene copolymers (ETFE), polychlorotrifluoroethylene (CTFE), tetrafluoroethylene-hexafluoropropylene-perfluoroalkyl vinyl ether copolymer (EPA), tetrafluoroethylene-hexafluoropropylene copolymers (FEP), tetrafluoroethylene-perfluoroalkyl vinyl ether (PFA),
15 polytetrafluoroethylene (PTFE), etc. Among them, tetrafluoroethylene base copolymers are preferable in view of the non-stick property. The fluororesins are preferably used in the form of an aqueous dispersion in view of the dispersibility.

The composition of the present invention can be applied and
20 cured in the same manners as those used to apply and cure the conventional curable compositions of the fluorine-containing copolymers. That is, according to the properties of the compositions, they are applied to an article to be coated by brush coating, spray coating, dip coating, flow coating, dispenser
25 coating, screen coating, etc., and sufficiently dried, followed by baking at a temperature of 150 to 300°C for 10 to 120 minutes.

The surface of the article to be coated is preferably defatted or washed. It is also preferable to form a primer layer

on the surface of the article to be coated to improve the adhesion of the composition to the article. Examples of the primer include silane primers, silicone primers, etc.

A surface layer may be formed on the coating film formed from the composition of the present invention. The surface layer can be formed from the above fluororesins and/or the terminal-modified perfluoropolyethers by conventional methods.

The curable composition of the fluorine-containing elastic copolymer according to the present invention is characterized in that it has better shelf stability than an aqueous composition curable with a polyamine curing agent, although the composition of the present invention is a one-fluid type aqueous dispersion. Furthermore, the composition of the present invention can contain the solid of the fluorine-containing copolymer at a higher concentration than the solvent-based polyol-curing composition.

Examples of articles or substrates to be coated with the composition of the present invention are as follows:

metals (e.g. iron, stainless steel, copper, aluminum, brass, etc.), glass products (e.g. glass plates, woven and non-woven fabric of glass fiber, etc.), molded articles or coated articles of general resins or heat-resistant resins (e.g. polypropylene, polyoxymethylene, polyimide, polyamideimide, polysulfone, polyethersulfone, polyether ether ketone, etc.), molded articles or coated articles of general rubbers (e.g. styrene-butadiene rubber (SBR), isobutylene-isoprene rubber, nitrile-butadiene rubber (NBR), styrene-propylene rubber (EPDM), etc.) and heat-resistant rubbers (e.g. silicone rubber, fluoroelastomer, etc.), woven and non-woven fabric of natural and synthetic fibers,

and the like.

The coating layers formed from the composition of the present invention can be used in various fields which require heat resistance, solvent resistance, lubrication and/or non-stick properties. Specific examples of the applications include rolls (e.g. fixing rolls, press rolls, etc.) and conveying belts for OA equipment such as copying machines, printers, facsimiles, etc.; sheets and belts; O-rings, diaphragms, chemical-resistant tubes, fuel hoses, valve seals, gaskets for chemical plants, engine gaskets, and the like.

EXAMPLES

The present invention will be illustrated by the following examples.

Example 1

Preparation of Pigment Paste A

A filler (MT carbon black) (20 wt. parts) and an acid-acceptor (MA-150 manufactured by KYOWA CHEMICAL INDUSTRIES, Ltd.; main component: MgO) (3 wt. parts) were dispersed in pure water (48 wt. parts) together with a surfactant (HS-208 manufactured by NFO Corporation; 20 % aqueous solution; main component: $C_8H_{17}-p-Ph-O-(CH_2CH_2O)_n-OH$ ($n=8$)) (2 wt. parts) to obtain a paste, which is referred to as "Pigment Paste A".

Preparation of coating composition

Pigment Paste A (73 wt. parts) and a surfactant (ADEKANOL UH-1498 manufactured by ASAHI DENKA KOGYO Co., Ltd.; main component: polymeric nonionic surfactant) (1.5 wt. parts) were added to the dispersion of a fluoroelastomer (DAIEL G-501 AN manufactured by Daikin Industries Ltd.; vinylidene fluoride-

tetrafluoroethylene-hexafluoropropylene copolymer) (100 wt. parts of the solid component), and well dispersed. Then, to this aqueous dispersion, the sodium salt of bisphenol AF (2.2 wt. parts) as a basic polyol curing agent and the octylate salt of DBU (1,8-diaza-bicyclo[5.4.0]-7-undecene) (U-CAT SA102 manufactured by SAN-APRO Co., Ltd.) (0.5 wt. part) as a curing accelerator were added to obtain a coating composition.

With the obtained coating composition, the stability of the composition and the mechanical property of the coating film were measured by the following methods:

Stability

The coating composition was poured in a polyethylene bottle and kept standing at 25°C. After 2 days, 7 days, 2 weeks, 1 month and 2 months, the state of the coating composition was observed.

Mechanical properties

The coating composition was poured in a metal vat and dried at room temperature for 5 days and then at a temperature of 80 to 100°C for 2 days, followed by baking at 200°C for 60 minutes. The formed film was peeled off from the vat. Then, a JIS No. 5 dumbbell-shaped sample was punched out from the film and subjected to the tensile test at a pulling rate of 500 mm/min.

Example 2

A coating composition was prepared in the same manner as in Example 1 except that sodium salt of hydroquinone was used in place of the sodium salt of bisphenol AF.

Example 3

A coating composition was prepared in the same manner as in Example 1 except that a phenolic resin soluble in an alkaline

water (DKK-1 manufactured by ASAHI ORGANIC MATERIAL INDUSTRIES, Ltd.) was used in place of the sodium salt of bisphenol AF.

Example 4

A coating composition was prepared in the same manner as in Example 1 except that DAIEL G-701 BP dispersion (manufactured by Daikin Industries Ltd.; vinylidene fluoride-hexafluoropropylene copolymer dispersion) was used in place of the DAIEL G-501 AN dispersion.

Example 5

A coating composition was prepared in the same manner as in Example 1 except that DAIEL G-701 BP dispersion (manufactured by Daikin Industries Ltd.; vinylidene fluoride-hexafluoropropylene copolymer dispersion) was used in place of the DAIEL G-501 AN dispersion, and sodium salt of hydroquinone was used in place of the sodium salt of bisphenol AF.

Example 6

A coating composition was prepared in the same manner as in Example 1 except that DAIEL G-701 BP dispersion (manufactured by Daikin Industries Ltd.; the dispersion of vinylidene fluoride-hexafluoropropylene copolymer) was used in place of the DAIEL G-501 AN dispersion, and a phenolic resin soluble in an alkaline water (DKK-1 manufactured by ASAHI ORGANIC MATERIAL INDUSTRIES, Ltd.) was used in place of the sodium salt of bisphenol AF.

Comparative Example 1

A coating composition was prepared in the same manner as in Example 1 except that bisphenol A was used in place of the sodium salt of bisphenol AF.

Comparative Example 2

A coating composition was prepared in the same manner as in Example 1 except that a polyamine curing agent (EPOMATE F-100 manufactured by YUKA SHELL Co., Ltd.) (2 wt. parts) and a silane coupling agent (A-1100 manufactured by Nippon Unicar Co., Ltd.) (9 wt. parts) were used in place of the polyol curing agent and the curing accelerator, respectively, and pure water (7.5 wt. parts) was used.

The compositions and the results of the experiments in Examples 1-6 and Comparative Examples 1-2 are summarized in Table 1.

Table 1

Component (wt. %)	Ex. 1	Ex. 2	Ex. 3	Ex. 4	Ex. 5	Ex. 6	C. Ex. 1	C. Ex. 2
Fluoroelastomer dispersion: -DAIEL G-501AN -DAIEL G-701BP	100	100	100	100	100	100	100	100
Filler: MT-carbon black	20	20	20	20	20	20	20	20
Acid-acceptor: MA-150	3	3	3	3	3	3	3	3
Surfactant: ADEKANOL UH-140S	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Polyol curing agent: -Na salt of bisphenol AF -Na salt of hydroquinone -DKK-1* -Bisphenol A	2.2	2.2	2.2	2.2	2.2	2.2	2.2	2.2
Curing accelerator: SA-102	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Polyamine curing agent: Epomate E-100	---	---	---	---	---	---	---	2
Silane coupling agent: A-1100	---	---	---	---	---	---	---	9
Stability	Gelled after 7 days	No change after 3 months	Gelled after 2 weeks	Gelled after 7 days	No change after 3 months	No change after 3 months	Immediately gelled	Gelled in 2 days
100% Modulus (kgf/cm ²)	13	14	20	15	12	23	---	69
Tensile strength (kgf/cm ²)	67	57	68	43	40	69	---	110
Elongation (%)	590	820	780	570	1000	740	---	240

* Phenolic resin soluble in alkaline water.

Example 7

With the coating film formed from a coating composition, which was prepared in the same manner as in Example 3, the adhesion property, non-stick property and tensile property was measured as follows:

Adhesion properties

The coating composition was spray coated on a substrate, well dried at 80 to 100°C and baked at 200°C for 60 minutes.

On the surface of the coating film, crosshatches (100 squares) were formed according to JIS K 5400-1990, 8.5.2, and an adhesive tape (manufactured by NICHIBAN) was closely adhered to the cross-hatched surface, and immediately peeled off. Using fresh adhesive tapes, the peeling was repeated 10 times, and the number of the remaining squares was counted.

As the substrates, an aluminum plate (A-1050), a polyimide film, a silicone rubber sheet and a fluoroelastomer sheet were used.

The aluminum plate was beforehand sand blasted, and coated with a silane compound (LORD Chemlok Y-4310; 10 wt. % aqueous solution) as a primer. The silicone rubber sheet was also coated with GLP-103SR (manufactured by Daikin Industries Ltd.; main component: silicone resin) as a primer.

Non-stick property

On the surface of the coating film, which was prepared in the same manner as in the test of the adhesion properties, one drop of pure water or n-cetane was dropped, and a contact angle was measured with a goniometer (manufactured by KYOWA KAIMEN KAGAKU KABUSHIKIKAISHA).

Mechanical property

The mechanical property was measured in the same manner as in Example 1.

Example 8

- 5 The adhesion properties, non-stick property and mechanical property was measured in the same manner as in Example 7 except that a coating composition, which was prepared in the same manner as in Example 6, was used.

Comparative Example 3

- 10 The adhesion properties, non-stick property and mechanical property was measured in the same manner as in Example 7 except that a coating composition, which was prepared in the same manner as in Comparative Example 1, was used.

Comparative Example 4

- 15 The adhesion properties, non-stick property and mechanical property was measured in the same manner as in Example 7 except that a solvent-based polyol curable fluoroelastomer coating composition (a vinylidene fluoride-hexafluoropropylene copolymer base coating composition manufactured by Daikin Industries Ltd.) was used as a coating composition, and the coating composition was coated on the substrate with a bar coater to form the coating film for the measurement of the adhesion properties and non-stick property.

Example 9

- 25 Preparation of Pigment Paste B

A filler (TALOX R-516L manufactured by TITANIUM INDUSTRIES, Ltd.; main component: Fe_2O_3) (3 wt. parts) and an acid-acceptor (MA-150 manufactured by KYOWA CHEMICAL INDUSTRIES, Ltd.) (3 wt.

parts) were dispersed in pure water (43 wt. parts) together with a surfactant (HS-208 manufactured by NFO Corporation; 20 % aqueous solution) (2 wt. parts) to obtain a paste, which is referred to as "Pigment Paste B".

5 Preparation of coating composition

Pigment Paste B (51 wt. parts) and a surfactant (ADEKANOL UH-140S manufactured by ASAHI DENKA KOGYO Co., Ltd.) (3.5 wt. parts) were added to the dispersion of a fluoroelastomer (DAIEL G-501 AN manufactured by Daikin Industries Ltd.; vinylidene fluoride-tetrafluoroethylene-hexafluoropropylene copolymer) (100 wt. parts of the solid component), and a FEP dispersion (manufactured by Daikin Industries Ltd.; solid content: 52 wt. %) (192 wt. parts) and a surfactant (NONION DS-60HN manufactured by NFO Corporation) were added, and the mixture was well dispersed.

10

Then, to this aqueous dispersion, an alkaline water-soluble phenolic resin (DKK-1) (2.2 wt. parts) as a basic polyol curing agent and the octylate salt of DBU (1,8-diaza-bicyclo-[5.4.0]-7-undecene) (U-CAT SA102 manufactured by SAN-APRO Co., Ltd.) (0.5 wt. part) as a curing accelerator were added to obtain

15

a coating composition.

20

With a coating film formed from the obtained coating composition, the adhesion properties, non-stick property and mechanical property was measured as follows:

Adhesion properties and non-stick property

25 The coating composition was spray coated on a substrate, well dried at 80 to 100°C and baked at 300°C for 15 minutes. Then, the adhesion properties and non-stick property was measured in the same manners as those described in Example 7.

Mechanical property

The coating composition was spray coated on an aluminum foil, well dried at 80 to 100°C and baked at 300°C for 15 minutes. The aluminum foil was dissolved with hydrochloric acid. Then, a JIS
5 No. 4 dumbbell-shaped sample was punched out from the film and subjected to the tensile test at a pulling rate of 500 mm/min.

Example 10

A coating composition was prepared in the same manner as in Example 9 except that DAIEL G-701 BP dispersion was used in
10 place of DAIEL G-501 AN dispersion, and the adhesion properties, non-stick property and mechanical property of the coating film were measured.

Comparative Example 5

The procedures of Example 9 were repeated except that a
15 FEP-added aqueous polyamine curable fluoroelastomer coating (a vinylidene fluoride-tetrafluoroethylene-hexafluoropropylene copolymer base coating composition; ratio of fluoroelastomer to fluororesin (solid content) = 1:1) cured with the diamine agent to obtain a coating composition, and the adhesion properties,
20 non-stick property and mechanical property of the coating film were measured.

Example 11

The coating composition prepared in Example 9 was spray coated on the substrate and well dried at 80 to 100°C. Then, a
25 FEP powder coating (manufactured by Daikin Industries Ltd.) was coated on the dried film and then baked at 300°C for 15 hours. The adhesion properties and non-stick property of the coating film were measured in the same manners as in Example 9.

The results of the measurements in Examples 9-11 and Comparative Examples 3-5 are summarized in Table 2.

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Table 2

Properties	Ex. 7	Ex. 8	C.E. 3	C.Ex. 4	Ex. 9	Ex. 10	C.Ex. 5	Ex. 11
Adhesion properties (crosshatch test)								
Substrate:								
-Aluminum	100/100	100/100	100/100	100/100	100/100	100/100	100/100	100/100
-Polyimide	100/100	100/100	100/100	100/100	100/100	100/100	100/100	100/100
-Silicone rubber	100/100	100/100	100/100	100/100	100/100	100/100	100/100	100/100
-Fluororubber	100/100	100/100	100/100	100/100	100/100	100/100	100/100	100/100
Non-stick property (contact angle)								
-Pure water	95	97	93	96	105	106	104	106
-n-Cetane	40	44	39	41	55	55	51	55
Mechanical properties								
-100% Modulus (kgf/cm ²)	20	23	69	40	55	60	120	---
-Tensile strength (kgf/cm ²)	68	69	110	90	85	85	140	---
-Elongation (%)	780	740	240	290	320	290	210	---

1 Amended pages under PCT Article 34

CLAIMS

1. (Amended) An aqueous fluoroelastomer curable coating composition comprising an aqueous fluoroelastomer dispersion and a basic polyol curing agent.

5 2. (Amended) The aqueous fluoroelastomer curable coating composition according to claim 1, wherein said basic polyol curing agent is a salt of a polyol having a phenyl group and at least two hydroxyl group with a basic compound.

10 3. (Amended) The aqueous fluoroelastomer curable coating composition according to claim 1, wherein said basic polyol curing agent is a salt of a polyol having a phenyl group and at least two hydroxyl group with at least one basic compound selected from the group consisting of ammonium, phosphonium, alkali metals and alkaline earth
15 metals.

4. (Amended) The aqueous fluoroelastomer curable coating composition according to claim 1, wherein said basic polyol curing agent is a salt of at least one polyol selected from the group consisting of hydroquinone,
20 bisphenol A, bisphenol AF and resol type phenolic resins and at least one basic compound selected from the group consisting of ammonium, phosphonium, alkali metals and alkaline earth metal.

25 5. (Amended) The aqueous fluoroelastomer curable coating composition according to claim 1, wherein said

fluoroelastomer is an elastic copolymer of vinylidene fluoride and at least one other ethylenically unsaturated fluorine-containing monomer comonomerizable therewith.

5 6. (Amended) The aqueous fluoroelastomer curable coating composition according to claim 1, which further comprises a fluororesin.

7. (Amended) The aqueous fluoroelastomer curable coating composition according to claim 6, wherein said fluororesin is a tetrafluoroethylene base polymer.

10 8. (Amended) The aqueous fluoroelastomer curable coating composition according to claim 6, wherein said fluororesin is a copolymer of tetrafluoroethylene with at least one monomer selected from the group consisting of hexafluoropropylene and perfluoroalkyl vinyl ethers.

15 9. (Amended) The aqueous fluoroelastomer curable coating composition according to any one of claims 1 to 8, which further comprises a terminal-modified perfluoropolyether.

20 10. (Amended) An article at least a part of the surface of which is coated with a coating layer formed from the aqueous fluoroelastomer curable coating composition according to any one of claims 1 to 9.

25 11. The article according to claim 10, wherein said coating layer is further coated with a non-stick layer of a fluororesin.

ABSTRACT

An aqueous fluoroelastomer curable composition comprising an aqueous fluoroelastomer dispersion and a basic polyol curing agent, and an article having a coating film formed from this composition. The aqueous fluoroelastomer curable composition has a long pot life although it is a one-fluid type aqueous dispersion, and forms a cured film having sufficient strength and heat resistance at a relatively low temperature in a short time.

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AQUEOUS. FLUOROELASTOMER CURABLE COMPOSITION AND COATED ARTICLE

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